Hot electrons in a tunnel structure based on metal nanoclusters

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We study the effect of temperature on the tunnel current in a structure based on gold clusters taking into consideration their discrete electronic spectra. We suggest that an overheating of electron subsystem leads to the disappearance of a current gap and gradual smoothing of current–voltage curves that is observed experimentally.

The nanodispersed metallic systems are prospective object of nanotechnology [1, 2, 3]. Transport of electrical charge across a nanoscale tunnel junction is accompanied by many effects, such as the Coulomb blockade of the average current, transfer of energy between electrons and ions, and consequent heating of the junction. In nanometer scale devices, electron transport can occur through well-resolved quantum states. If the temperature is increased, the Coulomb and quantum staircases of current are gradually smeared out by thermal fluctuations.

Simple tunnel construction can be schematically represented by the distinctive "sandwich" [1, 3]. It consists of a thick gold film (emitter) covered by a dielectric one (with dielectric constant $\epsilon \approx 3$). Disc-shaped [1] or spherical-like [3] gold clusters are self-organized on the dielectric layer. Also, a tip of STM is used in the capacity of the third electrode (collector).

Some of the experimental features of the $I-V$ curves were investigated in Ref. [4], however, the fact of smoothing of staircases for granule-molecule at low temperatures is still not understood. Such a smoothing is typical for molecular transistors [5]. Moreover, the observed current gap decreases significantly as temperature increases from 5 K to 300 K in structure based on disk-shaped cluster (Figure 2 in [1] for disk $2R \approx 4\,\mathrm{nm}$).

In this letter, the temperature features of the $I-V$ curves are explained by overheating of electron subsystem.

The number of atoms $N_0 \simeq \{14,\, 10^3\}$ and $\{100,\, 600\}$ correspond to gold discs of monatomic thickness and spheres whose radii vary in the range $2R \simeq \{1,\, 8.5\}$ and $\{1.4,\, 2.8\}\,\mathrm{nm}$, respectively. For given cluster sizes, the condition $L \gg R$ is fulfilled, where L is the free path length for the electrons in the bulk of a metal.

The calculation of the electron spectrum in the cylindrical and spherical wells of the mentioned sizes with finite deepness yields different values for the spectrum discreteness in magic clusters $\Delta\varepsilon_p = \varepsilon^{\mathrm{LU}} - \varepsilon^{\mathrm{HO}}$ (see Figure 1). In the nonmagic clusters the levels of lowest unoccupied states coincide with those of highest occupied ones, $\varepsilon^{\mathrm{LU}} = \varepsilon^{\mathrm{HO}}$ at $T = 0$.

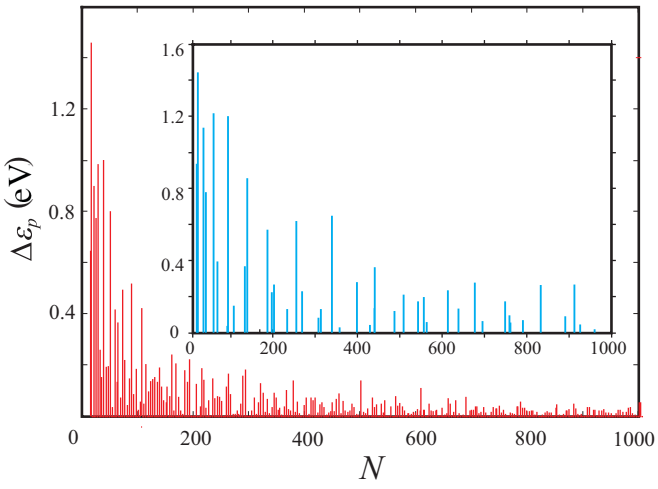

FIG. 1: Inset shows the specific difference between energies of lowest unoccupied electron state $\varepsilon^{\mathrm{LU}}$ and highest occupied one $\varepsilon^{\mathrm{HO}}$ in neutral discs (red) and spheres (blue) Au_{N_0} at $T = 0$.

The characteristic Coulomb energy of charging is $\widetilde{E}_{\mathrm{C}} = e^2/C$, where C is self-capacitance of single granule in a vacuum (in the case of a disc, the capacitance can be estimated as for the oblate spheroids of equal volume). The calculations of Ref. [4] demonstrated that these C are too small for the width of the current gap to be explained. The most obvious example is the case of a disc, since almost half of the disc surface contacts to the dielectric film. Therefore, for these granules we change $C \Rightarrow (1+\epsilon)C/2$. Then, for discs and spheres we have $\widetilde{E}_{\mathrm{C}} \simeq \{1.60,\, 0.21\}$ and $\{1.82,\, 1.06\}\,\mathrm{eV}$, respectively. We note that the value of the capacitance is sensitive to the shape of the granule surface, and even small deviation from the spherical shape can change significantly the capacitance.

The consequence of the phonon spectrum deformation of granules is the weakening of the electron-phonon interaction within them: $v_{\mathrm{F}}/R \gg \omega_{\mathrm{D}}$, where v_{F} is the electron velocity at the Fermi surface in the bulk, and ω_{D} is the Debye frequency. This interaction can be so suppressed that the electron-electron interaction becomes the main mechanism for the dissipation of the energy, which is in-

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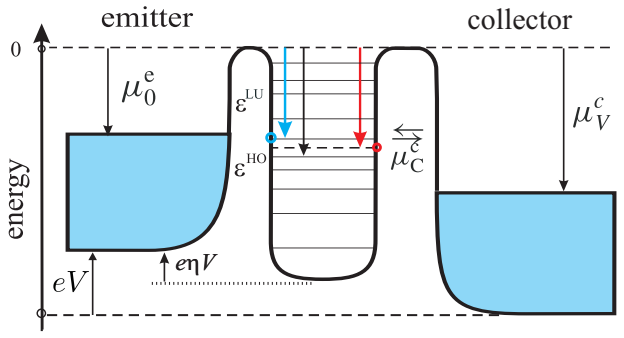

FIG. 2: Energy profile of structure for $V > 0$. μ_C^c is the electron chemical potential of charged granule in external electric field [4].

jected to the particle. This additional energy results in the overheating of the electron subsystem, which is described by the Fermi statistics with some effective (enhanced) temperature $T_{\rm eff}^{\rm g}$, and the temperature of the ion subsystem only slightly changes [6]. With the increase of the bias voltage V, the number of electrons, relaxing in the granule, increases significantly.

Among them are all the electrons with energies in the interval $e\eta V$ (see Figure 2) below the Fermi level of the granule (ηV is the fraction of the bias voltage on the granule), since the "flow" of tunneling electrons increases from below lying levels, thereby, involving large number of conductivity electrons to the relaxation process. At the same time, channels of losses appear, which are related to the generation of holes on the occupied levels and their subsequent recombination. The granule does not fragmentize at the significant overheating of the electron subsystem, because the $I-V$ curves are reproduced at the cyclic changes of the bias voltage [1, 3].

The estimate of the energy, which is pumped by the conductivity electrons to the granules of discontinuous films, is given in Ref. [7] ($\sim$ 0.2, 0.3 eV). This means that the experiments [1, 3] correspond to the Coulomb blockade regime in the region of current gap at the whole diapason of R and reasonable values of $T_{\rm eff}^{\rm g}$. Also, the quantum ladder can be smeared out by the thermal fluctuations,

$$\widetilde{E}_{\rm C} > \Delta\varepsilon_{\rm F} \gtrsim k_{\rm B}T^{\rm e,c,g},$$

where $\Delta\varepsilon_{\rm F}$ is the difference between discrete levels in the vicinity of the granule Fermi level, and $\Delta\varepsilon_{\rm F} = \Delta\varepsilon_p$ for magic clusters at $T = 0$.

According to the simple model of Ref. [4], we represent the emitter and the collector as the electron reservoirs with continuum spectrums, which are occupied in accordance with the Fermi-Dirac distribution with chemical electron potential $\mu_0^{\rm e,c} < 0$ and temperatures $T^{\rm e,c}$ equal to the thermostat one. In all cases the energy is counted off from the vacuum level. The electron chemical potential $\mu^{\rm g}$ of a granule in a quantum case can be defined by

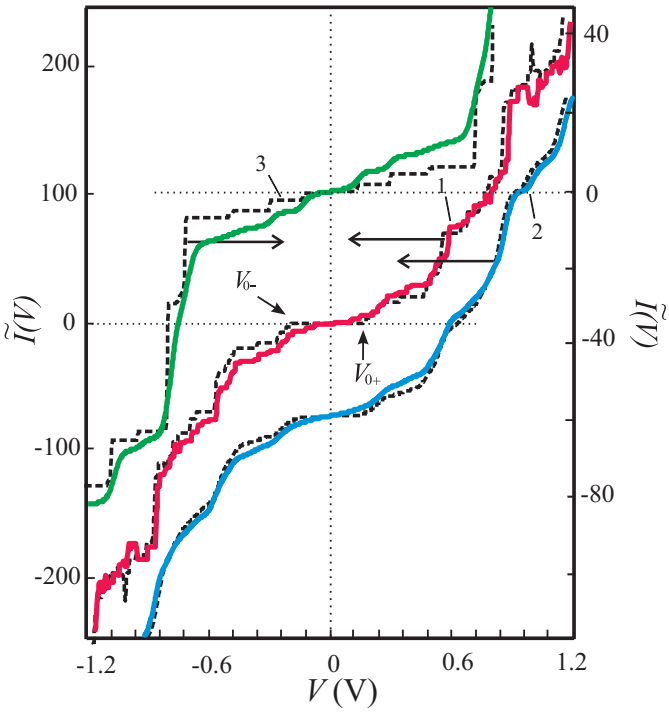

FIG. 3: Calculated $I-V$ curves of structure based on magic clusters: disc Au_{230} and sphere Au_{256}. 1 – Au_{230}: dotted curve – $T^{\rm e,c,g} = 5$ K, solid (red) – $T^{\rm e,c} = 5$ and $T_{\rm eff}^{\rm g} = 2000$ K. 2 – Au_{230}: dotted curve – $T^{\rm e,c,g} = 300$ K, solid (blue) – $T^{\rm e,c} = 300$ and $T_{\rm eff}^{\rm g} = 2000$ K. 3 – Au_{256}: dotted curve – $T^{\rm e,c} = 30$ K and $T_{\rm eff}^{\rm g} = 2000$ K, solid (green) – $T_{\rm eff}^{\rm e,c} = 300$ and $T_{\rm eff}^{\rm g} = 2000$ K.

the normalization condition at a given temperature $T_{\rm eff}^{\rm g}$:

$$\sum_{p=1}^{\infty}\{1+\exp[(\varepsilon_p-\mu^{\rm g})/k_{\rm B}T_{\rm eff}^{\rm g}]\}^{-1} = N_0, \qquad (1)$$

The summation in (1) is performed for all one-particle states, N_0 is the average number of conduction electrons in a granule. The spectrum of states is calculated in advance and, therefore, the chemical potential of neutral Au_{N_0} granules and its temperature dependence can be found from equation (1).

The current flowing through a metallic quantum granule (with restriction on its Coulomb instability [4]), is determined as $I^{\rm e} = I^{\rm c}$ or

$$-e\sum_{n_{\rm min}}^{n_{\rm max}} P_n\left(\overrightarrow{\omega_n^{\rm e}}-\overleftarrow{\omega_n^{\rm e}}\right) = -e\sum_{n_{\rm min}}^{n_{\rm max}} P_n\left(\overrightarrow{\omega_n^{\rm c}}-\overleftarrow{\omega_n^{\rm c}}\right), \qquad (2)$$

where P_n (the probability of the finding "in average" of n surplus electrons at the granule) is defined by the master equation in the stationary limit. In fact, the reduced current is calculated $\widetilde{I} \equiv I/(eP_0\Gamma^{\rm e})$, where $\Gamma^{\rm e,c}$ are the tunnel rates. In order to find values of $P_{n\neq 0}/P_0$ we use the recurrent relation. $\overleftrightarrows{\omega_n}$ are the partial electron "streams" from the last electrodes to the granule and in the opposite direction. For the comparison with the results of Ref.

[1, 3], the calculations are done for three temperatures of the collector and emitter $T^{\rm e} = T^{\rm c} = 5,\ 30,\ 300\,{\rm K}$, and also $T^{\rm g}_{\rm eff} = T^{\rm e},\ 2000\,{\rm K}$. The values $\Gamma^{\rm c} = \Gamma^{\rm e} = 1$ and $\eta = 1/2$ are used for all cases.

Figure 3 shows calculated $I-V$ curves for disc of radius $R = 2\,{\rm nm}$ (magic cluster Au_{230}) and sphere of radius $R = 1\,{\rm nm}$ (magic cluster Au_{256}). For low temperatures ($k_{\rm B}T^{\rm g}_{\rm eff} \ll \Delta\varepsilon_{\rm F}$), the current gap width $\Delta V_g = |V_{0-}| + V_{0+}$ is determined analytically by the conductance gap boundaries V_{0-} and V_{0+}. For example, V_{0+} is defined from the condition of absence of collector current of the direct $I-V$ curve branche ($V > 0$), and finally we have:

$$\Delta V_g = \Big(\frac{1}{2e}\tilde{E}_{\rm C} + \frac{1}{e}\Delta\varepsilon\Big)\Big[\frac{1}{2-\eta} + \frac{1}{1+\eta}\Big], \qquad (3)$$

where $\Delta\varepsilon \equiv \mu^{\rm g} - \varepsilon^{\rm HO} \geq 0$ at $T = 0$ and η is fixed as for $V > 0$. Calculated values of ΔV_g are in a good agreement with the experimental values based both on spherical and disc-shape clusters.

The calculation of the $I - V$ curves and current gap can be done only numerically at $k_{\rm B}T^{\rm g}_{\rm eff} \geq \Delta\varepsilon_{\rm F}$, when the larger part of the spectrum, compared to $\Delta\varepsilon_{\rm F}$, is responsible for the charge transfer. Our calculations show an evident dependence of $I - V$ curves flatness on the electron subsystem temperature.

However, it order to obtain an agreement with observed $I - V$ curves it is necessary to suggest that electrons in the emitter and collector are also heated up to some effective temperature, which is higher than the thermostat one. It is possible, because electrons (the current $I = 1\,{\rm pA}$ is provided by $I/e \sim 10^6$ number of electrons per second) relax in generally on the free path length in the last electrodes.

For the illustration, we present our result at Figure 3 for the sphere at $T^{\rm e,c}_{\rm eff} = 300\,{\rm K}$ and $T^{\rm g}_{\rm eff} = 2000\,{\rm K}$. Only by such a way, we can explain the flattening of the $I - V$ curves for the metallic cluster structures at low thermostat temperatures. With the increase of the bias voltage, the current flow is accompanied by the increase of the electron gas temperature.

In conclusion, we have calculated the $I - V$ characteristics of structure based on magic clusters: disc Au_{230} and sphere Au_{256}. We have suggested that the overheating of electron subsystem leads to the disappearance of current gap and significant flattening of current–voltage curves. Our results are in a good qualitative agreement with experiment of Ref. [1, 3].

Acknowledgments

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